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Evidence for long-range transport of carbon monoxide in the Southern Hemisphere from SCIAMACHY observations

A. M. S. Gloudemans,[1] M. C. Krol,[1,2,3] J. F. Meirink,[3] A. T. J. de Laat,[1,4]
G. R. van der Werf,[5] H. Schrijver,[1] M. M. P. van den Broek,[1] and I. Aben[1]

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[1] The SCIAMACHY satellite instrument shows enhanced carbon monoxide (CO) columns in the Southern Hemisphere during the local Spring. Chemistry-transport model simulations using the new GFEDv2 biomass-burning emission database show a similar temporal and spatial CO distribution, indicating that the observed enhancements are mainly due to biomass burning (BB). Large differences between the year 2003 and 2004 are observed in both the measurements and the model for South America and Australia. This study analyzes the origin of these observed enhancements in the Southern Hemisphere. The fact that SCIAMACHY is sensitive to surface CO allows for the observation of enhanced CO columns in both emission areas and in areas that are affected by long-range transport of CO. Model results show a large contribution of South American BB CO over Australian BB regions during the 2004 BB season of up to ~30–35% and up to 55% further south, with smaller contributions for 2003. BB CO transported from southern Africa contributes up to ~40% in 2003 and ~30% in 2004. The results indicate that differences between SCIAMACHY CO and the model simulations over Australian BB areas are probably not only caused by uncertainties in local emissions but also in overseas emissions. **Citation:** Gloudemans, A. M. S., M. C. Krol, J. F. Meirink, A. T. J. de Laat, G. R. van der Werf, H. Schrijver, M. M. P. van den Broek, and I. Aben (2006), Evidence for long-range transport of carbon monoxide in the Southern Hemisphere from SCIAMACHY observations, *Geophys. Res. Lett.*, *33*, L16807, doi:10.1029/2006GL026804.

1. Introduction

[2] With a lifetime of weeks to months carbon monoxide (CO) is an excellent tracer of atmospheric transport processes. It is also the major sink of OH and an important pollutant. One of its major sources is seasonal biomass burning (hereafter: BB). The SCanning Imaging Absorption spectroMeter for Atmospheric CHartographY (SCIAMACHY) on board the ENVISAT satellite, allows the measurement of global distributions of CO down to the Earth's surface. SCIAMACHY observations show significant CO total column enhancements over well-known BB areas [e.g., *Buchwitz et al.*, 2004, 2005; *Frankenberg et al.*, 2005; *Buchwitz et al.*, 2006; *de Laat et al.*, 2006]. Evidence for intercontinental transport of CO in the Southern Hemisphere (SH) has been found from several aircraft campaigns [e.g., *Staudt et al.*, 2001, 2002; *Pak et al.*, 2003; *Sinha et al.*, 2004], focussing on remote locations and short time periods of <6 months. Longer time series have been investigated by *Bowman* [2006] using trajectory analyses and qualitative comparison with MOPITT (Measurements Of Pollutants In The Troposphere [*Deeter et al.*, 2003]).

[3] Although near-infrared SCIAMACHY observations over sea are complicated due to low signal-to-noise, they do have a good sensitivity to surface CO over land contrary to thermal measurements as currently performed by MOPITT, AIRS (Atmospheric InfraRed Sounder [*McMillan et al.*, 2005]), and TES (Tropospheric Emission Spectrometer [*Beer et al.*, 2001]). Thus, both local emissions and CO transported from overseas emission areas can be studied. This allows to determine their relative contributions over emission areas as well as over more remote regions on a global scale for long time periods. Here a quantitative analysis of a 16 month period of SCIAMACHY CO data at 12 locations in the SH is presented. The study focusses on the origin of the observed CO enhancements over BB regions and nearby areas, as well as their interannual variation.

2. Measurements and Transport Model

[4] The SCIAMACHY CO total columns have been retrieved with the Iterative Maximum Likelihood Method (IMLM) version 6.3 between 2324.5–2337.9 nm [*Gloudemans et al.*, 2005; *de Laat et al.*, 2006]. The continuous growth of an ice layer on the detector and an increasing number of radiation-damaged detector pixels over time complicate the near-infrared retrievals. In-flight decontaminations, an improved calibration, and inclusion of an ice layer correction in the IMLM retrievals have reduced their effects [*Gloudemans et al.*, 2005]. *de Laat et al.* [2006] show that the resulting SCIAMACHY CO total columns agree well spatially and temporally with chemistry-transport model simulations when using monthly mean CO on a $3^\circ \times 2^\circ$ lon-lat grid, provided the signal-to-noise ratio of the spectra and the number of collocations are sufficient. Here, the same data set, selection criteria, and weighting procedure as in *de Laat et al.* [2006] is used to construct monthly mean CO total columns on a $3^\circ \times 2^\circ$ lon-lat grid. Whereas *de Laat et al.* [2006] only use SCIAMACHY observations with cloud cover <20%, here also data with collocated CH_4 columns $>3 \times 10^{19}$ molec/cm^2, corresponding to clouds

[1]Netherlands Institute for Space Research, Utrecht, Netherlands.
[2]Meteorology and Air Quality Group, Wageningen University, Wageningen, Netherlands.
[3]Institute for Marine and Atmospheric Research, Utrecht University, Utrecht, Netherlands.
[4]Royal Netherlands Meteorological Institute, de Bilt, Netherlands.
[5]Faculty of Earth and Life Sciences, Free University, Amsterdam, Netherlands.

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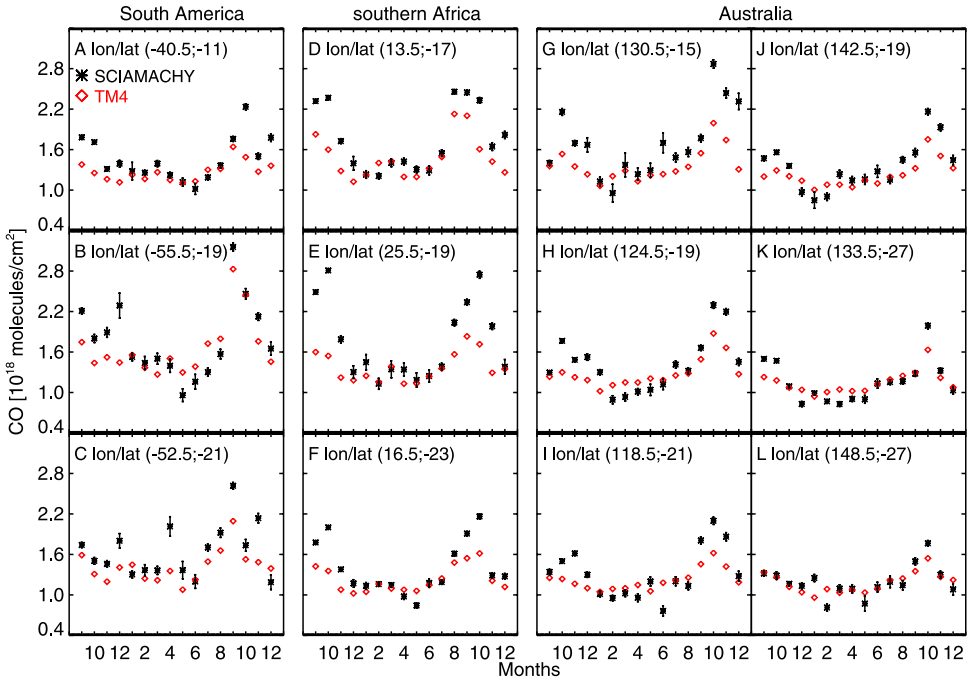

Figure 1. Comparison of SCIAMACHY (black) and TM4 (red) monthly mean variability for 12 SH locations (see Figure 2) between September 2003 and December 2004. Error bars denote the 1σ noise errors.

and/or smoke from forest fires below 1.5 km are included in order to increase the number of collocations over BB areas. Although in this case only the CO above the smoke/cloud is measured, no significant bias is found compared to *de Laat et al.* [2006], suggesting that these observations contain smoke/clouds well below 1.5 km.

[5] The model calculations are performed with the same global chemistry-transport model (TM4) as in *de Laat et al.* [2006] on a 3° × 2° lon-lat grid. However, the independent satellite-based BB emission database (GFEDv2) from *van der Werf et al.* [2006] is used rather than the climatological emissions from the EDGAR 2.0 database [*van Aardenne et al.*, 2001] as used by *de Laat et al.* [2006]. The vertical distribution of the BB emissions follows the recommendation for the AEROCOM model intercomparison [*Dentener et al.*, 2006]. In addition, so-called tagged tracer simulations have been performed with a transport-only version. In these simulations a BB CO tracer was released over either South America, Africa, Australia, or Indonesia + Southeast Asia. Chemical destruction of CO in that version is calculated by using monthly averaged OH fields from the full chemistry simulation while retaining monthly mean diurnal variation in OH.

3. SCIAMACHY Compared to Model CO

[6] Figure 1 shows the comparison between monthly mean SCIAMACHY CO and the temporally and spatially collocated TM4 results at 12 SH 3° × 2° grid boxes for the period September 2003–December 2004. Locations near (A, F, K, and L in Figure 2) and over (B–E and G–J in Figure 2) BB emission areas are selected in order to capture both local emission and transport of CO. At all locations CO columns are available for all 16 months with a mean monthly random noise error better than ∼6% based on up to 47 collocations per month (see auxiliary material[1] Table S1), warranting a reliable seasonality of the data. Such a precision is achieved over areas with a moderate to high surface albedo in the near infrared [*de Laat et al.*, 2006]. Systematic retrieval errors over such regions are estimated to be below 10%.

[7] Outside the BB season, model and SCIAMACHY agree well with an average difference <5%, much smaller than the bias *Dils et al.* [2006] find for SH ground-based comparisons. However, the ground-based comparisons cover only 3 SH stations with few exact collocations, surface albedos <0.1, and SCIAMACHY random noise errors well above 10%. During the 2004 BB season, i.e., about August to November, South America and Australia show much larger CO enhancements than in 2003, with higher values in South America than in Australia. In southern Africa the 2003 and 2004 maxima seem comparable. The interannual variations in the modeled CO columns are mainly driven by the BB CO emissions as described by *van der Werf et al.* [2006] and only to a minor extend by transport differences, since simulations with climatological emissions used in *de Laat et al.* [2006] show only minor interannual variability in modeled CO. The resulting model enhancements around September–October are in good agreement with the observed SCIAMACHY CO columns. The GFEDv2 emissions by *van der Werf et al.* [2006], which are based on actual fire counts scaled to burned area, thus provide a major improvement. The MOPITT CO columns over South-America shown in *Bowman* [2006] hardly vary between 2003 and 2004. MOPITT, however, has lower sensitivity to the boundary layer, which contains most of the BB CO in emission areas.

[1] Auxiliary material data sets are available at ftp://ftp.agu.org/apend/gl/2006gl026804. Other auxiliary material are in the HTML.

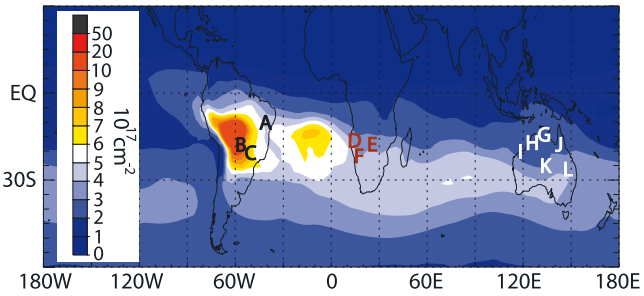

Figure 2. CO column results for October 2004 from the tracer model including only BB emissions in South America. Transport of CO from South America to Southern Africa and Australia is clearly visible. Locations A–L from Figure 1 are shown.

[8] Figure 1 also shows significant differences between model and SCIAMACHY. For instance, the observed enhancements are generally larger than the model simulations. These differences are significant compared to those found outside the BB season. Possible biases in the retrievals or due to modeling assumptions are also much smaller. This indicates that, although the timing of the BB emissions seems correct in the GFEDv2 database, the absolute magnitude may be too low. CO emissions in South America may be high because of deforestation as well as maintenance fires, but frequent cloud cover hinders satellite observation of these fires. Therefore, the satellite-based GFEDv2 emissions probably underestimate real emissions [*van der Werf et al.*, 2006]. In December 2003 locations B and C show much larger SCIAMACHY CO columns compared to the model. Within a month, the GFEDv2 CO emissions are evenly distributed in the model. However, the MODIS fire counts are about 6 times higher in the second week of December 2003 than in the first week [*Giglio*, 2005]. Valid SCIAMACHY CO columns are available only for this second week, which may explain the higher SCIAMACHY values.

[9] In southern-African location E the SCIAMACHY CO peaks ~1 month later than the model. *Van der Werf et al.* [2006] note that in southern Africa the fire season shifts through time from west (D, grassland) to east (E, woodland) and that greater burned area in the west and their corresponding emissions combined with their coarse spatial resolution modeling framework leads to a peak somewhat too early for regions east of ~25°E. The SCIAMACHY CO columns seem to confirm this, since E maximizes later than the more westerly location D. In addition, CO emissions from woodland burning (E) may be underestimated in the GFEDv2 database, because the low spatial resolution hampers a differentiation between grassland and woodland fires [*van der Werf et al.*, 2006]. Location F is located south of the emission region and transport probably causes the later CO BB maximum in both the model and SCIAMACHY.

4. Origin of CO Excess

[10] The Australian locations are particularly interesting since long-range transport from South America and Africa has been shown to affect CO concentrations over remote areas in the South Pacific and over southeast Australia [*Staudt et al.*, 2002; *Pak et al.*, 2003]. The CO columns over mid Australia (K and L) maximize around October, whereas in South America the BB peak occurs around September. Southern Africa maximizes around September–October depending on the location. The analysis presented by *Bowman* [2006] shows that South-American emissions reach Australia within a few weeks, in good agreement with the SCIAMACHY observations. His analyzed wind fields for October 2002 suggest that long-range transport could also have a pronounced effect over the northern BB emission areas in Australia (G–J) and that both southern African and Indonesian emissions may contribute as well.

[11] The individual contributions for 2003 and 2004 are quantified using the tracer model described in section 2. The rationale is that interannual variability in CO emissions is dominated by BB rather than other CO sources. Figure 2 shows that the high South American CO emissions in 2004 lead to accumulation over the southern Atlantic. Part of this accumulated CO enters the westerlies and is transported to Australia. Analyses of Cape Grim surface measurements show no enhancements, confirming that the CO transported from South America is located in the free troposphere (auxiliary material Figure S1) [*Pak et al.*, 2003].

[12] Figure 3 shows the CO contribution of the different tracers and their sum. The SCIAMACHY BB CO has been estimated by subtracting the modeled non-BB columns from the total observed columns. The results confirm the qualitative analyses by *Bowman* [2006] and show that in mid-Australia (K,L) the CO columns are dominated by BB CO transported from South America during the 2004 BB season. During the 2003 BB season, South America and Africa each contribute ~35–40%. *Sinha et al.* [2004] find comparable contributions for South America and Africa in 2000. This agrees well with the southern-Africa-to-South America BB emission ratio, which is about equal in 2000 and 2003 [*van der Werf et al.*, 2006]. African 2004 emissions are comparable to 2003. In contrast, South American BB emissions in 2004 are about 2–3 times larger than in 2003 [*van der Werf et al.*, 2006]. This agrees well with the 2004/2003 CO column ratios of the South American BB tracer over Australia (G–L) of 2.2–3.1 and over South American emission regions of 2.6–3.1 (Figure 3, purple lines). Thus, the interannual variability of the South American CO column contributions over Australia are largely determined by the 2003–2004 emission variations over South America. This strengthens our findings that the 2003–2004 interannual variation in CO columns over Australia is strongly related to variability in South American BB emissions and only to a minor extend to transport variability. Interestingly, the contribution of Australian fires during the BB season is <27% over mid-Australia (K,L). According to the GFEDv2 inventory, even in northern Australia (I,J) these fires are not the major contributor. At G and H which are in the center of the fire region, the Australian contribution dominates, but even here the South American emissions contribute ~25–30% in 2004 and ~15–30% in 2003. An important fraction of the enhanced CO columns observed by SCIAMACHY over Australia is thus due to intercontinental transport of CO emitted in South America.

[13] In addition to the findings over Australia, the contribution of South American emissions to the CO excess

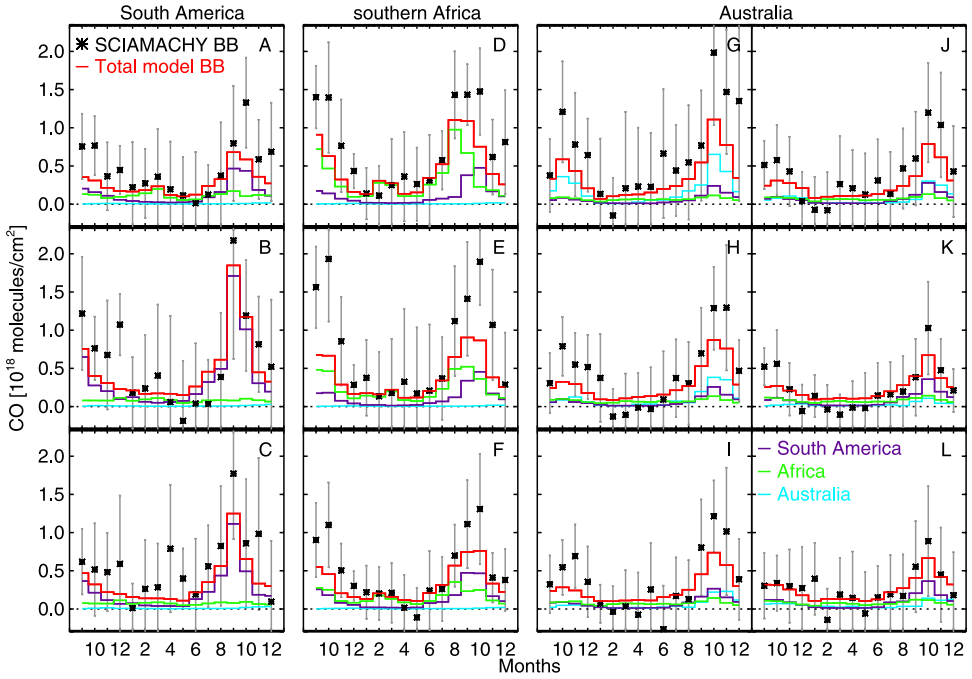

Figure 3. Contribution of different CO BB sources to the SCIAMACHY CO columns at locations A–L, as derived from the tracer model, which is sampled in the same way as SCIAMACHY CO. Indonesia + Southeast Asia contributes $<0.17 \times 10^{18}$ molecules/cm^2 and is therefore not shown. Corresponding numbers of all tracers are listed in Table S2 of the auxiliary material. The SCIAMACHY BB CO is estimated by subtracting the modeled non-BB columns from the total observed columns. The 1σ monthly variability of SCIAMACHY (grey bars) - which is larger than the noise error - is mostly larger than the modeled variability (auxiliary material Table S2) because the model has a lower spatial resolution (3° × 2°) and contains only monthly mean emissions. The black dashed line shows the zero level.

over Africa (D–F) is also found to be significantly higher in 2004 than in 2003, emphasizing that the interannual variability of South American BB emissions has a profound effect on observed CO columns over all three SH continents.

5. Concluding Remarks

[14] The findings reported here indicate that differences between model simulations and SCIAMACHY observations cannot be explained by systematic errors in SCIAMACHY CO or uncertainties in the local emission estimates only. Intercontinental transport of BB CO, for instance from South America to Australia, clearly contributes to the observed CO columns. Furthermore, significant interannual variation in the contributions of local and overseas BB CO has been identified, which is largely due to interannual variability in emissions rather than transport variability. The sensitivity of SCIAMACHY to surface CO provides good opportunities for the inverse estimation of CO emissions. Moreover, synergy with MOPITT, AIRS, and TES which are mostly sensitive to middle and upper tropospheric CO, will allow a better quantification of the contribution of local and overseas emissions in order to improve current emission estimates.

[15] **Acknowledgments.** SCIAMACHY is a joint project of the German Space Agency DLR and the Dutch Space Agency NIVR with contribution of the Belgian Space Agency. We thank the Netherlands SCIAMACHY Data Center and ESA for providing data. The work performed is (partly) financed by the EC (5th Framework Programme, project EVERGREEN) and NIVR.

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I. Aben, A. T. J. de Laat, A. M. S. Gloudemans, M. C. Krol, H. Schrijver, and M. M. P. van den Broek, SRON, Sorbonnelaan 2, NL-3584 CA Utrecht, Netherlands. (a.gloudemans@sron.nl)

J. F. Meirink, IMAU, Utrecht University, Princetonplein 5, NL-3584 CC Utrecht, Netherlands.

G. R. van der Werf, Faculty of Earth and Life Sciences, Free University, De Boelelaan 1085, NL-1081 Amsterdam, Netherlands.